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Electromigration experiments for studying transport parameters and sorption of cesium and strontium on intact crystalline rock

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ABSTRACT

This study aims to determine upscaling factors for the radionuclides' distribution coefficients (K_d) on crushed rocks to intact rock for the safety analysis of radionuclide migration from spent nuclear fuel in bedrock towards biosphere. Here we report the distribution coefficients for intact rock determined by electromigration sorption experiments and compare the results with those from recently performed batch sorption experiments. In total 34 rock samples, representing three typical rock types from Olkiluoto Finland, were studied in order to determine distribution coefficients, effective diffusion coefficients and porosities using the electromigration sorption experiments, formation factor experiments and porosity measurement. The parameters determined represent the three main parameters of geosphere used in the safety assessment of spent nuclear fuel disposal. The distribution coefficients of cesium and strontium on intact rock varied between $(0.12–26.2) \times 10^{-3}$ m^3/kg and $(1.4–13.3) \times 10^{-3}$ m^3/kg, respectively, whereas recent results for crushed rock varied between $(2–57) \times 10^{-3}$ m^3/kg and $(17–40) \times 10^{-3}$ m^3/kg, respectively. This implies that crushing increases the distribution coefficient significantly and upscaling factors from 3 to 33 were determined for scaling the distribution coefficients of crushed rock to ones of intact rock. The determined distribution coefficients of cesium and strontium for intact rock can be directly applied in the safety assessment whereas the upscaling factors can be used to convert distribution coefficients determined for crushed rock into ones for intact rock. Based on the results for porosities and effective diffusion coefficients it was concluded that they do not seem to correlate with sorption parameters. However, an alteration state, heterogeneity and mineral content seem to be important factors affecting the distribution coefficients and upscaling factors.

1. Introduction

In Finland, the spent nuclear fuel from the currently operating nuclear power reactors will be disposed of in a bedrock repository at a depth of about 400 m. In the final disposal of spent nuclear fuel in geological formations, such as bedrock in Finland, the rock above the waste acts as the last barrier against the release of radionuclides into the biosphere. This has been taken into account in the safety assessment of the final disposal concept by the evaluation of the transport properties of radionuclides in the bedrock. The retardation mechanisms of radionuclides in bedrock are matrix diffusion and sorption on the minerals (Posiva, 2013). In the safety assessment effective porosity, diffusion coefficient and distribution coefficient (K_d) define the transport properties of the rock, and the magnitude of matrix diffusion and sorption. The effective diffusion coefficient depends only slightly on the considered radionuclide while the distribution coefficient varies strongly from radionuclide to another. Furthermore, the transport parameters, especially the distribution coefficient, depend on the rock type and the mineral composition of rock. The porosity is fairly straightforward to determine while the distribution and effective diffusion coefficient offer challenges, particularly in case of crystalline rock. The distribution coefficients are typically determined using crushed rock. This, however, creates new surface area (directly in contact with water molecules) capable of sorbing radionuclides and thus distribution coefficients determined using crushed rock samples overestimate the distribution coefficient of intact rock. It has been found out that the distribution coefficients determined using crushed rock can be significantly higher than the ones determined using intact rock (Crawford, 2010). Furthermore, the batch sorption experiments are typically performed using a water-rock ratios that are not realistic for intact rocks. The diffusion coefficients, on the other hand, are typically measured using long-lasting and tedious experiments.

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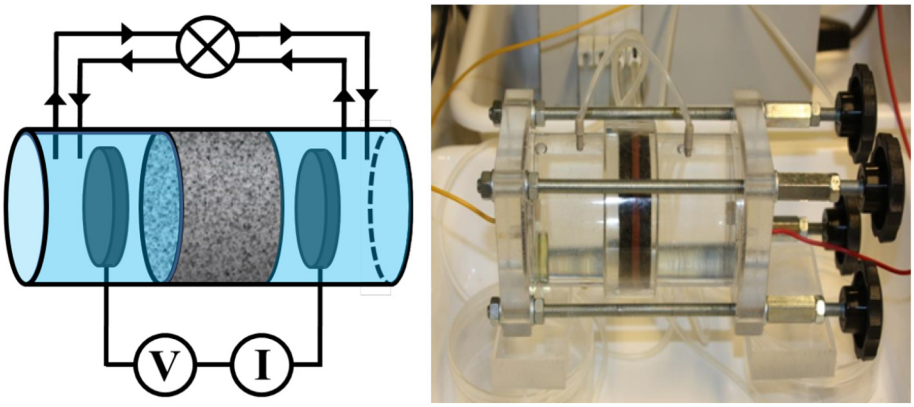

Fig. 2. *A schematic diagram of the electromigration equipment (left)* that consists a *voltage source and meter (V), ammeter (I), peristaltic pump (X), platinum electrodes and drill core sample and a photograph of the electromigration experimental vessel (right) showing the drill core samples in the middle of two chambers.*

assumed that pore space of the rock affects the effective diffusion coefficient and resistivity of the rock similarly. This assumption leads to F_f being equivalent with the pore geometry factor (G) that commonly defined to be a function of porosity and tortuosity. In our experiments there were Cl^- and Na^+ ions in the solution and their diffusion coefficients in water at room temperature are $2.0 \times 10^{-9}\,m^2/s$ and $1.3 \times 10^{-9}\,m^2/s$, respectively. Their average value ($1.65 \times 10^{-9}\,m^2/s$) was used when determining the effective diffusion coefficients.

3. Results

3.1. Porosities, formation factors and e ective di usion coe cients

The porosities, formation factors and effective diffusion coefficients of the drill core samples used in the electromigration sorption experiments are presented in Table 2. The observed porosity values represent the typical values for rock samples taken from the Olkiluoto area (Smellie et al., 2014). However, these values are closer to the low porous end of typical values. There was no correlation found between the determined porosities and rock type that is typical for crystalline rock samples taken from the same site.

The results for F_f and D_e are given as an uncertainty weighted averages over all independent measurements. Here different electric potentials and their uncertainties were taken into account. The variations of the results over each drill core and rock type arise from the heterogeneity of the rock and, more importantly, the heterogeneity of the pore structure. Such variations are common in crystalline rock at the studied scale (Sammaljärvi et al., 2017).

Table 2
The average volumetric porosities, formation factors (F_f) and effective diffusion coefficients (D_e) of rock samples used in the electromigration sorption experiments (a number of core samples permeasurements in parentheses). The values are given as an uncertainty weighted average over all individual experiments and given uncertainties are uncertainties of the weighted average. Uncertainty of individual core samples varied between 20% and 50% for porosity, between 4% and 21% for the formation factor and between 4% and 20y% for the effective diffusion coefficient.

Rock Type	Sample (number of samples)	Porosity (%)	$F_f \times 10^{-5}$ (−)	$D_e \times 10^{-14}$ (m^2/s)
Mica gneiss (MGN)	PP219 (6)	0.26 ± 0.05	14 ± 3	24 ± 6
	PP309 (6)	0.07 ± 0.01	4 ± 2	7 ± 3
	PP131(51) (3)	0.36 ± 0.02	n.d.	n.d.
	MGN average	0.23 ± 0.12	11 ± 8	18 ± 13
Tonalite-granodiorite-granite-gneiss (TGG)	PP175 (7)	0.07 ± 0.01	3 ± 2	6 ± 4
	KR56 (3)	0.19 ± 0.08	38 ± 7	64 ± 11
	KR43 (2)	0.22 ± 0.01	n.d.	n.d.
	TGG average	0.15 ± 0.08	13 ± 17	22 ± 29
Pegmatitic granite (PGR)	PP131(70) (2)	0.46 ± 0.06	n.d.	n.d.
	PP249 (3)	0.19 ± 0.01	n.d.	n.d.
	PGR average	0.30 ± 0.15	n.d.	n.d.

n.d. = not determined.

The average effective diffusion coefficients of MGN samples were $(24 \pm 6) \times 10^{-14}\,m^2/s$ (PP219) and $(7 \pm 3) \times 10^{-14}\,m^2/s$ (PP309). The difference is relatively small when taking into account the differences in mineral composition, grain size and foliation. It can be concluded that slightly larger grain size (probably lower tortuosity of pore space) and more heterogeneous structure of the PP219 drill core has caused its slightly larger D_e compared with one of PP309 drill core. These values are also in accordance with the porosities (PP219: 0.24 ± 0.04%; PP309: 0.07 ± 0.01%). Typically a sample with higher porosity has a higher D_e than a sample with lower porosity if the pore structure remains similar.

The average effective diffusion coefficients for TGG samples were $(6 \pm 4) \times 10^{-14}\,m^2/s$ (PP175) and $(64 \pm 11) \times 10^{-14}\,m^2/s$ (KR56). The difference is over one order of magnitude and it reflects large differences in mineral composition and in pore structure between these rock samples. Examination of the mineral compositions reveal (see Table 1) that the PP175 samples have much higher content of biotite than the KR56 samples. The pore space of biotite consists of space between the randomly ordered biotite grains and nanometer scale apertures between the biotite lamellae within each grain. The nanometer scale pores have extremely low constrictivity and they tend to decrease the D_e of the rock if biotite content is high. In this case straight conclusion according to porosity (PP175: 0.07 ± 0.01%; KR56: 0.19 ± 0.08%) and D_e cannot be drawn due to the remarkable difference in mineral content and pore structure. However, the porosities happen to be larger for the samples from the drill core KR56 than the ones from the drill core PP175. For all of the samples determined effective diffusion coefficients are within the range of ones determined in earlier studies (Smellie et al., 2014). However, the values are closer to the high diffusivity end of typical values.

3.2. Distribution coe cients for intact rock and their up scaling factors

The electromigration sorption experiments were done to determine the distribution coefficients of cesium and strontium for intact rock and to determine the upscaling factors for distribution coefficients from crushed to intact rock. Seven example curves for the concentration of cesium in the solution chambers as a function of time are shown in Fig. 3. These concentration curves represent the rate of sorption: larger change indicates greater sorption and a larger distribution coefficient. The experiments were run until a steady state concentration was reached. Typically, the equilibrium was reached after 2000–7000 h (see Fig. 3). The equilibrium concentration was then used for determining

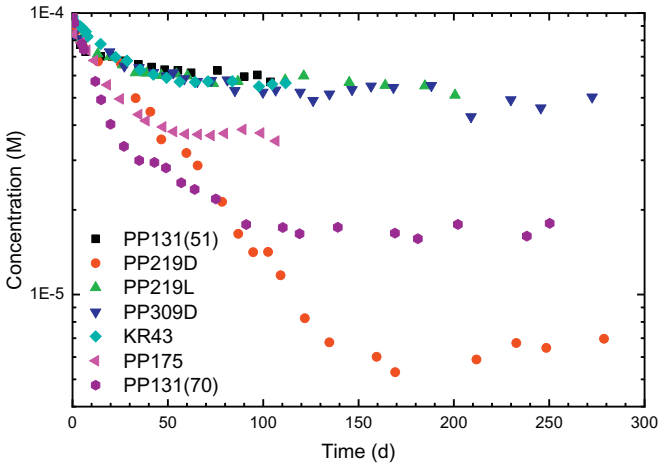

Fig. 3. Concentration of cesium as a function of time in seven out twenty-eight electromigration sorption experiments. Initial cesium concentration was 10^{-4} M.

the distribution coefficients (see Table 3). It is obvious that porosity, K_d and F_f of the rock affect the shape of the concentration curve and the time when equilibrium is reached. The K_d determines the value where the concentration levels out. Because electric potential is used to transport cesium through the sample, and more importantly closer to the potential sorption sites, the F_f affects the curve part before leveling out. In practice, for samples with high F_f the concentration becomes constant faster than the ones with low F_f (e.g. see curve for PP131(70) in Fig. 3). For samples with a low F_f, it takes a long time to achieve the equilibrium concentration since it takes time to reach the sorption sites even with electric potential as driving force.

The distribution coefficients were determined from the steady state concentration using Eq. 1 and the results for all rock samples and averages for studied rock types are given in Table 3. The selection criteria for equilibrium concentration were as follows:

1. If the concentration had leveled, an average of the 2–3 last values was used.
2. If the concentration had an increasing trend at the end of the experiment, the average value of all points after an initial decrease was used
3. If the concentration was still decreasing at the end of the experiment, " < " sign was used to indicate that the steady state value is smaller than the presented one. This means that steady state value for distribution coefficients (crushed and intact) are greater than the presented ones. In this case the upscaling factor is given as most probable value and is indicated with "∼" sing.

The distribution coefficients of cesium for crushed rock were taken from curves presenting the distribution coefficients as a function of cesium concentration (Lehto et al., 2018) at the same cesium concentration as was the equilibrium concentration in the electromigration sorption experiment with the same rock sample. For the samples not studied by Lehto et al. (2018) (PP131(51), KR43, PP131(70) and PP249) the distribution coefficients were determined by separate experiments with equilibrium concentration of about 50 M that was a typical value for equilibrium concentration is these experiments.

The distribution coefficients of cesium for crushed rock are from 1.1 to 60 times greater than the ones of intact rock (see Table 3). In general, the batch sorption experiments exaggerate the distribution coefficient of intact rock considerably and the average upscaling factors of 7 ± 4, 3 ± 1 and 33 ± 30 were determined for MGN, TGG and PGR samples, respectively. The variances between different rock types, however, are

Table 3
The distribution coefficients (K_d) of cesium for intact Olkiluoto rocks from the electromigration sorption experiments and ones for crushed rocks at the same equilibrium concentration (Lehto et al., 2018). The upscaling factors (F_U) are given as a ratio of distribution coefficients of crushed and intact rocs. The average values are given as uncertainty weighted averages and initial cesium concentration was 10^{-4} M.

Sample	Rock type	Intact rock K_d (m^3/kg) 1×10^{-3}	Equilibrium Cs concentration (M)	Crushed rock K_d (m^3/kg) 1×10^{-3}	F_U (−)
PP131(51)_1	MGN	2.8 ± 0.5	53 ± 5	25 ± 8	9 ± 3
PP131(51)_2		11.4 ± 0.5	20 ± 1	36 ± 9	3.2 ± 0.8
PP131(51)_3		2.3 ± 0.2	59 ± 4	24 ± 6	10 ± 3
PP219A		27 ± 2	11 ± 1	46 ± 7	1.7 ± 0.3
PP219D		50 ± 6	6.2 ± 0.3	57 ± 9	1.1 ± 0.3
PP219H		1.8 ± 0.5	67 ± 13	22 ± 5	12 ± 5
PP219L		2.5 ± 0.4	58 ± 6	24 ± 4	9 ± 2
PP309A		> 4.0	< 48	> 16.3	∼4.1
PP309D		3.4 ± 0.3	49 ± 3	16.2 ± 1.3	4.8 ± 0.6
PP309H		1.2 ± 0.3	75 ± 13	14 ± 2	11.8 ± 4
PP309L		2.6 ± 0.1	57 ± 3	15.5 ± 1.1	6.0 ± 0.5
PP309C		1.8 ± 0.1	64 ± 9	15 ± 2	8.3 ± 1.1
PP309E		3.5 ± 0.1	44 ± 4	17 ± 2	4.8 ± 0.5
MGN average		**2.7 ± 0.6**		**17 ± 3**	**7 ± 4**
KR43_1	TGG	2.6 ± 0.2	42 ± 2	12.1 ± 1.1	4.7 ± 0.6
KR43_2		3.5 ± 0.3	52 ± 3	11.4 ± 1.0	3.3 ± 0.4
KR43_4		4.9 ± 0.1	51 ± 3	11.6 ± 1.0	2.4 ± 0.3
PP175A		4.7 ± 0.3	64 ± 4	12.9 ± 1.0	2.7 ± 0.3
PP175D		5.9 ± 0.3	68 ± 4	13.4 ± 1.0	2.3 ± 0.3
PP175V		4.1 ± 0.1	61 ± 3	12.6 ± 0.9	3.1 ± 0.8
KR56Q		> 0.7	< 81	> 2.1	∼3.1
KR56O		> 1.6	< 64	> 2.2	∼1.4
TGG average		**4 ± 2**		**10 ± 5**	**3 ± 1**
PP249_1	PGR	0.22 ± 0.05	90 ± 2	8.1 ± 0.7	37 ± 9
PP249_2		0.13 ± 0.02	94 ± 1	7.8 ± 0.7	60 ± 10
PP249_3		0.12 ± 0.05	95 ± 2	7.7 ± 0.7	60 ± 30
PP131(70)_11		17.7 ± 1.3	17 ± 1	27 ± 3	1.5 ± 0.2
PP131(70)_12		26.2 ± 1.1	11 ± 1	32 ± 4	1.2 ± 0.2
PGR average		**1 ± 11**		**30 ± 11**	**33 ± 30**

not very similar. For rock samples with a high biotite content the variation of the upscaling factors are rather similar whereas for PGR samples higher variation was observed. For the MGN samples the upscaling factors vary from 1.1 to 12, for the TGG samples they vary from 1.4 to 4.7, and for PGR samples they vary from 1.2 to 60. In general, the variations in the upscaling factors are rather high which might make it difficult to convert a distribution coefficient of single batch sorption to one for intact rock. For MGN and TGG samples the variation can be explained by heterogeneity in the porespace and mineral content of the rock samples that can be seen in Fig. 1. Furthermore, for samples of each drill core there are both low and high upscaling factors determined. Previously Kuva et al. (2018) have shown the effect of mineral heterogeneity on the transport of cesium in crystalline rock samples from Olkiluoto. For PGR samples the low upscaling factors were determined for samples of PP131(70) and the high upscaling factors for the ones from PP249. The same applies also for distribution coefficients determined for both intact and crushed rock. The PGR samples contain a few percent of biotite and variation in its content cannot explain this. However, it is plausible that the samples of the different drill core have undergone different alteration processes. Due to seritization and muscovization of plagioclase more reactive mineral grains and more surface area is created. Effect of this can be seen as a higher distribution coefficient of cesium for the samples of the drill core PP131 than the ones of the drill core PP249. When less altered drill core PP249 is crushed respectively more surface area is created and thus the upscaling factor becomes greater for less altered samples than for more altered ones. Lehto et al. (2018) observed that crushing does not only create more surface on the outer surfaces of the grains but also the inner surface due to mechanical stress. Proportion of inner and outer surfaces varied considerably from sample to another and grain size affected significantly proportion. It can be concluded that alteration state of the sample can affect considerably the distribution coefficient and the upscaling factor and thus a number of measurement using samples with different alteration states should be used in order to get a representative result for the average upscaling factor.

There was no clear correlation found between effective diffusion coefficient and distribution coefficient. It seems that pore structure of the rock affects more the effective diffusion coefficient, whereas the distribution coefficient and upscaling factor are more affected by the mineral content and alteration state.

The electromigration sorption experiments were done for strontium with drill cores PP219 (MGN) and PP175 (TGG). Example curves for the decrease of strontium concentration in the solution phase as a function of time are shown in Fig. 4. As in the cesium experiments, these

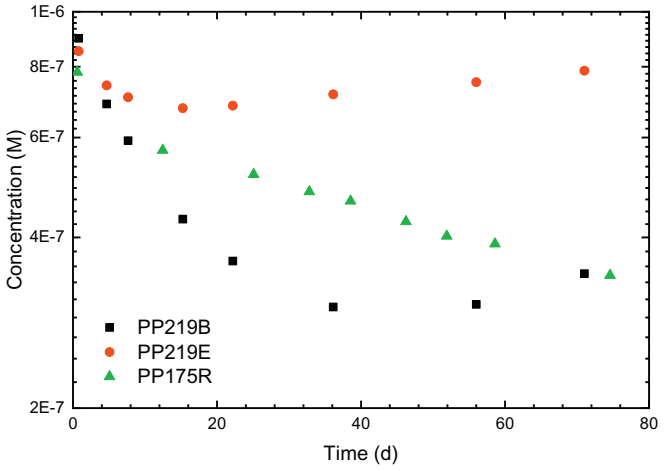

Fig. 4. Concentration of strontium in three out of six electromigration sorption experiments with PP219 (MGN) and PP175 (TGG) as a function of time. Initial strontium concentration was 10^{-6} M.

Table 4
The distribution coefficients of strontium (K_d) for intact Olkiluoto rocks from the electromigration sorption experiments and corresponding values for crushed rocks at the same equilibrium concentration. The upscaling factors (F_U) are given as a ratio of the distribution coefficients of crushed and intact rocs. The average values are given as uncertainty weighted averages and initial strontium concentration was 10^{-6} M.

Sample	Rock type	Intact rock K_d (m^3/kg) 1×10^{-3}	Equilibrium Sr concentration (M)	Crushed rock K_d (m^3/kg) 1×10^{-3}	F_U (−)
PP219B	MGN	7.4 ± 0.8	0.32 ± 0.02	17.6 ± 0.6	2.4 ± 0.3
PP219E		1.4 ± 0.3	0.72 ± 0.1	17.3 ± 0.6	12 ± 3
MGN average		**4.4 ± 3.0**		**17.4 ± 0.2**	**4 ± 3**
PP175A	TGG	> 13.3	< 0.14	40 ± **2**	< 3.0
PP175E		> 5.7	< 0.25	40 ± **2**	< 7.0
PP175R		> 6.3	< 0.34	40 ± **2**	< 6.4
PP175U		> 3.8	< 0.42	40 ± **2**	< 10.5
TGG average		**7 ± 3**		**40 ± 2**	**7 ± 2**

experiments were run until an equilibrium concentration was reached (typically 300–1400 h). The equilibrium concentration was then used for determining the distribution coefficients (see Table 4). The concentration of strontium was still decreasing at the end of the experiments with PP175 samples and thus the distribution coefficients are given as lower limits in Table 4. For drill core PP219 (MGN), the concentration starts to increase slightly after 20–60 days which is plausibly caused by small changes in water chemistry due to water-rock interaction. The effect of the concentration decrease has been, however, taken into account as higher uncertainty for equilibrium concentration. For strontium, the equilibrium concentration did not affect considerably the distribution coefficient for crushed rock at the studied concentration range (Lehto et al., 2018) and thus the maximum values for the upscaling factors of PP175 samples are given. The shape of the concentration curves and the value for leveling out were otherwise interpreted in the same way as for the cesium curves. The concentration curves for MGN samples leveled out faster than the ones for TGG samples which is caused by systematic difference in the measured formation factors and effective diffusion coefficients. The distribution coefficients of strontium for crushed rock have been measured by Lehto et al. (2018).

The values of the upscaling factors for strontium did not differ considerably from those of cesium with MGN and TGG (no experiments were done with PGR). Also for strontium the batch sorption experiments exaggerate the distribution coefficient of intact rock considerably and the average upscaling factors of 4 ± 3 and 7 ± 2 were determined for MGN and TGG, respectively (see Table 3). For the MGN samples the upscaling factors vary from 2.4 to 12, for the TGG samples they vary from 3 to 10.5. As in the measurements using cesium the variation in mineral content and heterogeneity can explain the variation of results within both rock types. Furthermore, there was no correlation found between the upscaling factor and effective diffusion coefficient. In general, there is no significant difference between the results of cesium and strontium and thus it can be concluded that variation in the mineral content and properties of the rock affect more the results than radionuclide in question. Here we have used cesium and strontium that both are sorbed on the mineral surfaces by ion exchange.

4. Discussion

The determined porosities and diffusion coefficients were within the range of ones determined in earlier studies. However, the values for effective diffusion coefficients were closer to the high diffusivity end of typical values while the porosities were closer to the low porosity end. These findings are in contradiction as, in general, effective diffusion coefficient increases with increasing porosity. Observed effect is likely

arising from the slight overestimation of the effective diffusion coefficients by the electrical formation factor measurements. Löfgren (2015) compared effective diffusion coefficients determined by through diffusion measurements and electrical formation factor measurements from earlier studies and found that the electrical methods tend to overestimate the effective diffusion coefficient by a factor of 1.2 to 2.4 potentially due to presence of electrically conductive mineral grains. The variation in the over estimation is fairly high and is clearly dependent on the content of the electrically conductive mineral grains within the transport routes. On the other hand, the content of these minerals must correlate with the rock type. In order to improve knowledge between overestimation and rock type further studies could be performed.

It was shown that the distribution coefficients for intact rock are considerably lower than the ones determined for crushed rock. However, the variations of the upscaling factors were found to be rather high which reduces their applicability when converting results of batch sorption experiments for intact rock. The upscaling factors give further insight which kind of values should be used when estimating the distribution coefficient of intact rock based on results for crushed rock as most of the distribution coefficients used in current safety assessment are determined by batch sorption experiments. However, in future studies it would be beneficial to use the electromigration sorption experiments for determining distribution coefficients in order to increase the confidence of the results. Currently the batch sorption experiments are needed for studying the sorption of redox sensitive radionuclides. In theory, it might be possible to study the sorption and diffusion of the redox sensitive radionuclides. This, however, would require further development of the measurement setup and experimental techniques due to, e.g., the effect of electrical voltage on the oxidation state.

5. Conclusions

The main aim of this work was to determine the distribution coefficients of intact rock and to produce the upscaling factors of distribution coefficients determined by batch sorption experiments to those of intact rock. To this end, the electromigration sorption experiments were performed here for intact samples and the upscaling factors were determined using the distribution coefficients determined for crushed rock samples by Lehto et al. (2018).

It was shown that the distribution coefficients of cesium and strontium for intact rock are considerably lower than the ones determined for crushed rock and thus it can be concluded that the electromigration sorption experiments offer a more realistic approach to determine the distribution coefficients of radionuclides for intact rock. The average upscaling factor for different rock types varied from 3 to 33 that shows large variation within rock types. These factors can be used to convert the distribution coefficients of cesium and strontium for crushed rock to ones for intact rock. It was concluded that heterogeneity of the rock, alteration state and mineral content affect the distribution coefficients and further the upscaling factors. The upscaling factors for cesium and strontium are very similar and the properties of the rock affect more the results than the radionuclide in question. In general, the sorption parameters determined here offer ways for a less conservative approach (with respect to the current approach) for the safety assessment of spent nuclear fuel repositories. Furthermore, the methods used here can be applied for other safety relevant anionic and cationic radionuclides.

The determined effective diffusion coefficients and porosities were in agreement with previously determined values for samples of Olkiluoto. The methods applied offer fast and reliable ways to measure these parameters before the electromigration sorption experiments without destroying the samples. In the end, all three parameters of bedrock used in the safety assessment of the spent nuclear fuel repositories were determined here for exactly the same samples. The values can be directly applied in the safety assessment and they can be used to increase its confidence and decrease the conservativeness of the values used current in safety assessment.

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